

(19)



**Europäisches Patentamt**  
**European Patent Office**  
**Office européen des brevets**

(11) Publication number:

**0 109 145**  
**A2**

(12)

**EUROPEAN PATENT APPLICATION**

(21) Application number: 83304982.8

(51) Int. Cl.<sup>3</sup>: **C 08 L 77/10, C 08 L 33/24,**  
**C 08 F 8/32**

(22) Date of filing: 30.08.83

(30) Priority: 10.11.82 US 440672

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(43) Date of publication of application: **23.05.84**  
**Bulletin 84/21**

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(84) Designated Contracting States: **AT BE CH DE FR GB IT**  
**LI LU NL SE**

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(54) **Compatible polymer blends of imidized acrylic polymer and polyamide and their use in the production of plastic articles.**

(57) **Polymer blends of 1 to 99% by weight of imidized acrylic polymer, containing at least 5% by weight of glutarimide units, and 99 to about 1% by weight of polyamide are disclosed which may possess attractive physical properties and may find use in the production of plastics articles.**

**EP 0 109 145 A2**

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COMPATIBLE POLYMER BLENDS OF IMIDIZED ACRYLIC POLYMER AND  
POLYAMIDE AND THEIR USE IN THE PRODUCTION OF PLASTICS  
ARTICLES.

5 This invention concerns compatible polymer blends of  
imidized acrylic polymer and polyamide and their use in the  
production of plastics articles.

10 The invention is more particularly concerned with  
compatible blends, of (a) polyglutarimide and (b) nylon,  
which can possess desirable characteristics of either (a) or  
(b).

15 In general, physical blending of polymers has not been  
a successful route for combining into a single material the  
desirable individual characteristics of two or more  
polymers. Typically, blends of polymers result in a  
material which combines the poorest characteristics of each  
polymer such that the resulting blend is not of any  
practical value. Thermodynamics, and particularly the  
entropy of mixing polymers, dictates that most polymer  
20 blends will not be miscible or compatible and will result in  
a material having two or more distinct phases, each  
containing predominantly only one of the starting polymers.  
Miscibility, as used herein, refers to the complete  
intermixing of the individual polymers in each other on a  
molecular level to form a solution. Compatibility, as used  
25 herein, is a more general term than miscibility but should  
not be confused with miscibility. Compatibility describes  
the ability of polymers to combine together for beneficial  
results. Compatible polymer blends appear macroscopically  
uniform, although on a molecular level the individual  
30 polymers may or may not be completely miscible in each other  
and may or may not form true polymer solutions.

In order to obtain combinations of polymers which  
exhibit the desirable characteristics of each polymer,  
copolymerization of the polymers or the incorporation of

compatibilizers which are soluble in each of the polymers have been employed. U.S. Patent No. 4,090,996; Mechanical Properties of Polypropylene - Low Density Polyethylene Blends, Nolley, Barlow and Paul, Polymer Engineering and Science, March 1980, Vol. 20, No. 5, p. 364; A Brief Review of Polymer Blend Technology, Paul and Barlow, Multiphase Polymers Cooper and Estes, Adv. Chemical Series 176 (1979); Block Copolymers, Allport and Janes, p. 365.

Polyglutarimides, including those disclosed in U.S. Patent No. 4,246,374 (Kopchik) possess good thermal stability, high service temperatures (°C Vicat), high tensile strength, solvent and hydrolysis resistance, good weatherability properties, low melt flow rates, and high melt strength. Polyglutarimides are, however, brittle and exhibit poor impact resistance. The impact resistance or toughness of polyglutarimides alone may be improved by the addition of conventional impact modifiers, for example, ABS (acrylonitrile/butadiene/styrene), MBS (methylmethacrylate/butadiene/styrene), all acrylic modifiers, polycarbonates, and mixtures thereof (U.S. Patent Nos. 4,217,424 and 4,254,232). However, it has been found that conventional polycarbonate-containing impact modifier systems for polyglutarimides are not miscible or compatible with polyglutarimides and can result in anisotropy in injection molded articles prepared therefrom. This injection molding induced anisotropy is caused by the melt flow orientation of insoluble, deformable inclusions of polycarbonate in the polyglutarimide. As a result, the impact modified polyglutarimide is tougher in the direction of flow into a mold than it is in the transverse direction. This anisotropy makes injection molded articles prepared from conventionally impact modified polyglutarimides containing polycarbonate unsuitable for certain applications requiring uniform toughness.

Despite the commercial utility of poly(carbon)imides, referred to herein as polyamides, for example in the

preparation of fibers, polyamides have been found to be unsuitable for most extrusion and blow molding processes because of their high melt flow rates and poor melt strength. Accordingly, attempts have been made to modify polyamides to achieve better strength characteristics by the incorporation of modifiers. U.S. Patents Nos. 3,668,274; 3,796,771; and 3,784,497 disclose modifications to nylon 6 and nylon 66 by incorporating acrylic core//shell polymers. These polymer additives improve the impact resistance and melt flow characteristics of the nylons but lower other important performance parameters, such as tensile strength and modulus, and result in opaque materials. Other patents relating to the modification of nylons include U.S. Patent No. 3,963,799 (graft reaction of polyamide and ethylene-based rubber modifier); U.S. Patent 4,035,438 (mixture of polycaprolactam and polyethylene and a graft polymer of ethylene/(meth) acrylic acid copolymer and grafted polymerized units of E-caprolactam); U.S. Patent 4,086,295 (incorporation of a copolymer of ethylene and a carbonate or epoxy compound); U.S. Patent 4,100,223 (incorporation of polyethylene and emulsifying copolymer of aromatic vinyl compound and an alpha,beta-unsaturated mono- and/or dicarboxylic acid or anhydride having free acid groups optionally neutralized by mono- and/or polyamides, ammonia, and esters of the mono- or dicarboxylic acid); U.S. Patent 4,167,505 (acrylic core//shell modifier dispersed in higher molecular weight nylon followed by blending with low molecular weight nylon); U.S. Patent 4,174,358 (polyamide matrix resin and branched or straight chain polymer of specified particle size range and tensile modulus); U.S. Patent 4,246,371 (polyamide with copolymer of ethylene, (meth)acrylic acid and esters thereof); U.S. Patent 4,293,662 (polyamide blended with oxidized polyethylene); U.S. Patent 4,321,336 (polyamide blended with either aliphatic polyolefin, olefinic copolymer of aliphatic monoolefin, (meth)acrylic acid or esters thereof, copolymers

of styrene, (meth)acrylic acid and esters thereof,  
copolymers of aliphatic dienes and acrylonitrile and high  
molecular weight organic silicon compounds); U.S. Patent  
4,336,406 (polyamide and graft product of polybutadiene and  
5     grated units of t-butyl acrylate and an acrylic acid ester  
with 1-4 carbon atoms in the alcohol moiety); and U.S.  
Patent 4,346,194 (polyamide blend for low temperature impact  
strength and ductility formed by adding elastomeric olefin  
copolymer or ionic copolymer of alpha-olefin and  
10     alpha,beta-unsaturated carboxylic acid and a third  
copolymerizable monomer such as an acrylate or  
methacrylate). There are, however, no reported attempts to  
achieve improved impact resistance and ductility for  
polyglutarimides by blending the polyglutarimide with  
15     polyamide, or for achieving improved melt flow, service  
temperature, toughness, and improved clarity for polyamides  
by blending polyamides with polyglutarimides.

It is an object of the present invention to provide a  
compatible polymer blend of polyglutarimide and polyamide  
20     such that the compatible polymer blend possesses certain  
desirable characteristics of each of the polymer components.

Accordingly the present invention seeks to provide a  
compatible polymer blend of polyglutarimide and  
fibre-forming polyamide (nylon) wherein the blend possesses  
25     improved melt flow, melt strength, and modulus as compared  
with the nylon alone.

The invention also seeks to provide a compatible  
polymer blend of low service temperature polyamide and  
higher service temperature polyglutarimide wherein the blend  
30     possesses improved service temperature than the low service  
temperature polyamide component.

It is an additional object of this invention to provide  
a compatible polymer blend of polyglutarimide and nylon 11  
wherein the compatible polymer blend possesses improved  
35     clarity compared to nylon 11.

It is a further object of the invention to provide a

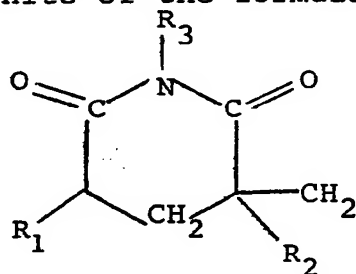
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compatible polymer blend of polyglutarimide and polyamide wherein the compatible polymer blend possesses improved impact resistance and ductility than the polyglutarimide component.

5 In accordance with the present invention, there is provided a compatible polymer blend of polyglutarimide and polyamide comprising 1 to about 99% by weight of polyglutarimide and 99 to 1% by weight of polyamide.

10 It has been unexpectedly found that imidized acrylic polymer containing at least 5% by weight of glutarimide units (referred to herein as polyglutarimide) and polyamide form compatible polymer blends over the weight ratio range of 99/1 to 1/99 polyglutarimide to polyamide, and that  
15 compatible polymer blends containing 5 to 95% by weight of polyglutarimide and 95 to 5% by weight of polyamide possess certain desirable physical properties not possessed by the poorer performing polymer component.

Polyglutarimide which forms compatible polymer blends with polyamide include, for example, the imidized acrylic  
20 polymer disclosed in U.S. Patent 4,246,374 (Kopchik). This imidized acrylic polymer is formed by the reaction between acrylic polymer containing units derived from esters of acrylic or methacrylic acid and ammonia or primary amines. Imidized acrylic polymer containing at least 5% by weight of  
25 glutarimide units of the formula (I)



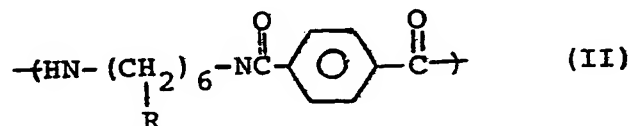
(I)

wherein  $R_1$ ,  $R_2$ , and  $R_3$ , independently, represent hydrogen or unsubstituted or substituted  $C_1$  or  $C_2$  alkyl, aryl, or aralkyl, or mixtures thereof, is referred to herein as polyglutarimide. Polyglutarimide derived from ammonia

imidization is referred to herein as 'ammonia imide' ( $R_3=H$ ) and polyglutarimide derived from imidization by methylamines are referred to herein as 'methyl imide' ( $R_3=CH_3$ ). The Vicat softening point of polyglutarimide as determined by ASTM DI 525-70, measured in degrees Centigrade, is used herein as a prefix to identify the polyglutarimide employed, i.e. '140° methyl imide'. The Vicat softening point, which is a measurement of the maximum service temperature of polyglutarimides, ranges from 125°C to 220°C.

Polyglutarimide found useful in this invention contain at least 5% by weight of glutarimide units and may be prepared by solution, emulsion, and autoclave processes as well as by melt reaction processes. The polyglutarimide useful in this invention may include other comonomer, such as styrene, in concentrations up to and including 95% by weight. The polyglutarimide is preferably non-crosslinked, soluble in dimethylformamide and thermally stable. The polyglutarimide may also be modified by the incorporation of additives, such as pigments, fillers, stabilizers, lubricants, and impact modifiers.

Polyamide found to form compatible polymer blends with polyglutarimide includes aliphatic and aromatic polyamide. Aliphatic polyamide includes polylactams, for example, nylon 6, nylon 11, and nylon 12, as well as polyhexamethylene adipamide (nylon 66). Aromatic polyamide includes poly(alkyl-substituted) hexamethylene diamine terephthalate having the structural formula (II)



wherein R is an alkyl group. An example of aromatic polyamide found to be compatible with polyglutarimide in Trogamide<sup>®</sup> T manufactured by the Kay Fries Company. Other aliphatic and aromatic polyamide and copolymers thereof may

also form compatible polymer blends with polyglutarimide.

The compatible polymer blends of this invention may be formed by any conventional melt or solvent blending technique; however, melt blending is preferred. The melt blends may be prepared in any device capable of melt mixing the polymers, such as extruders, heated mill rolls, sigma and Brabender mixers, etc. If an extruder device is used to melt mix the polymers to form a compatible polymer blend, a single screw extrusion device, for example, a Killion extruder or a twin screw extruder, for example a tangentially counter-rotating twin screw extruder manufactured by the Welding Engineers Company, may be utilized.

It has also been unexpectedly found that compatible polymer blends of polyamide and polyglutarimide possess certain desirable physical properties as compared with the polyamide component alone. Compatible polymer blends of polyamide and polyglutarimide exhibit improved service temperature properties than low service temperature polyamide, improved modulus and strength, and improved melt flow and melt strength while preserving most of the original toughness of the polyamide component alone.

When lower service temperature (DTUFL) polyamide, such as nylon 6, nylon 11, or nylon 66, is blended with at least 10% by weight, and preferably at least 25% by weight, of higher service temperature polyglutarimide, for example 160° Vicat methyl imide, the service temperature of the resulting compatible polymer blend is higher than the service temperature of the corresponding polyamide component. The service temperature of a compatible polymer blend and each of the polymer components may be determined by a distortion temperature under flexural load test (DTUFL) (ASTM 648). Generally, the DTUFL at 264 psi is about 20 to 30 C° lower than the Vicat softening temperature of polyglutarmides and is accurate to only plus or minus 5 C°.

In addition to the improvement in the service



temperature of the compatible polymer blends of low service temperature polyamide and higher service temperature polyglutarmide, these compatible blends, and particularly those blends containing at least 5% by weight and up to 75% by weight of nylon 11 and from 25% to 95% by weight of methyl imide, exhibit improved clarity compared to the corresponding polyamide component alone.

In addition to improving the service temperature and clarity of low service temperature polyamide, compatible polymer blends of polyamide and polyglutarimide exhibit significantly improved modulus or rigidity compared to the polyamide component. Polyglutarimide is rigid and has high moduli, whereas aliphatic polyamide is more flexible, less rigid, and has much lower moduli. By forming compatible polymer blends of polyamide and polyglutarimide, the modulus of the blend can be significantly improved compared to the polyamide component. This improvement in modulus as well as service temperature and clarity permits the use of compatible polymer blends of polyamide and polyglutarimide, and particularly compatible polymer blends of nylon 11 and polyglutarimide, in applications requiring clarity, toughness, and rigidity at service temperatures equivalent to, or significantly higher than, the service temperature of the polyamide component alone, depending on the ratio of imide to polyamide in the blend.

One of the most important aspects of this invention is that compatible polymer blends of polyamide and polyglutarimide exhibit significantly improved melt flow and corresponding melt strength properties than polyamide. It is well-known that unblended nylon melts have very low viscosity and are unacceptable for use in extrusion and other forming processes to prepare strands, sheets, films, and blow molded articles. The conventional addition of large amounts of impact modifiers to nylons to improve melt strength as well as impact resistance results in products having reduced moduli and poor optical properties (opaque).

The melt flow rate of unblended nylon 6 at 450° F and 400 psi is 195 grams per 10 minutes (g/10 min.), while at 525° F and 400 psi the melt flow rate increases to 478 g/10 min. The melt flow rate of unblended nylon 11 is even poorer than unblended nylon 6, being 320 g/10 min. at 450° F and 400 psi. The melt flow rate of nylon 66 is much lower than nylon 6 or nylon 11 and is 83 g/10 min. at 450° F; however, this is still too rapid for effective extrusion operations. Compatible polymer blends of polyamide and at least 5% by weight polyglutarimide exhibit reduced melt flow rates and correspondingly increased melt strength. These compatible polymer blends, containing up to 50% by weight of polyglutarimide, are acceptable for extrusion and other forming processes. As the weight ratio of the polyglutarimide to polyamide in the compatible polymer blend increases, the melt flow rate of the blend decreases and the melt strength increases. Accordingly, compatible polymer blends of polyamide and polyglutarimide can be prepared to achieve the desired melt rheology characteristics for a particular forming operation. These physical blends are simple to prepare and offer significant advantages over the addition of various modifiers, compatibilizers, and copolymers to neat polyamides.

Compatible polymer blends of polyglutarimide and polyamide comprising mixtures or copolymers of polyamide which exhibit the desired physical properties of the individual polymer components of the blend may also be prepared. In addition, the compatible polymer blends of this invention may include other modifiers or additives, for example, fillers, reinforcing agents, colors, pigments, flame retardants, processing aids, and impact modifiers, etc.

It has also been found that compatible polymer blends of polyglutarimide and polyamide possess certain improved physical properties compared to pure polyglutarimide and polyglutarimide modified with conventional impact modifiers.

Injection molded plaques of compatible polymer blends of polyglutarimide and polyamide possess improved toughness, as determined by a Gardner Vertical Height Impact Test (GVHIT), than the polyglutarimide component alone. The impact resistance of compatible blends of polyglutarimide and polyamide may be essentially equal to polyglutarimide modified with a butadiene-based impact modifier, plus an all acrylic impact modifier, plus a polycarbonate impact modifier (U.S. Patent 4,217,424, Ex. 9). However, injected molded articles formed from compatible polymer blends of polyglutarimide and nylon 11 are not subject to inclusion anisotropy or polycarbonate orientation effects as is the aforesaid conventional polycarbonate-containing impact modified polyglutarimide. Compression molded articles of the compatible polymer blends, therefore, can have the same degree of toughness as articles prepared by injection molding. Polycarbonate-containing impact modified polyglutarimide does not exhibit impact reinforcing anisotropy upon compression molding and thus may exhibit less than half of the impact resistance of corresponding injection molded samples. The preferred compatible polymer blend of polyglutarimide and polyamide which exhibits improved impact resistance or toughness over polyglutarimide alone and which does not exhibit inclusion anisotropy is a compatible polymer blend containing equal proportions of methyl imide and nylon 11.

It has also been found that compatible polymer blends of methyl imide and nylon 11 exhibit improved optical characteristics compared to impact modified polyglutarimide. Conventionally impact modified polyglutarimide provides opaque compositions while compatible polymer blends of methyl imide and nylon 11 are optically clear. Clarity, as used herein, refers to the ability of an observer to view objects through an article without distortion or loss of definition, i.e. glass. Transparency, as used herein, refers to the ability of an observer to view objects through

an article but with some loss of definition. Translucency, as used herein, refers to the ability of an article to transmit visual light, but the light is scattered by refraction and reflection and it is not possible to view objects through the article. Opaque, as used herein, refers to an article which does not transmit visual light therethrough. The optical properties reported herein are visually determined and the results are reported according to the above definitions. The preferred compatible imide/nylon polymer blends of this invention which exhibit clarity contain 5% to 75% by weight of nylon 11 and 95 to 25% by weight of 140° to 150° Vicat methyl imide. As the Vicat softening point of the methyl imide component of the compatible polymer blend increases to 160° Vicat and above, the compatible blend remains clear up to 50% by weight of nylon 11. At 60% by weight of nylon 11 and above, the blend begins to become hazy. Hazy is a condition somewhere between true clarity and transparency. The clarity and differential scanning calorimetry data of the preferred nylon 11/methyl imide blends indicate that these polymers may be truly miscible.

It has also been found that compatible blends of 140° to 180° Vicat methyl imide and nylon 6 or nylon 11 in a 95/5 weight ratio are also clear; however, compatible blends of 140° Vicat and 150° Vicat methyl imide and greater than 5% nylon 6 exhibit transparency and translucency.

The improved optical properties of the preferred methyl imide/nylon blends, and particularly those blends containing 5 to 50% by weight of nylon 11, permits these blends to be useful in applications requiring optical clarity and toughness, such as in skylights, glazing, signs, containers, and films.

Compatible blends of polyglutarimide and nylon can exhibit an improved response to the addition of conventional multi-stage impact modifiers than do polyglutarimide alone. For example, the incorporation of a two-stage acrylic impact

5 modifier comprising a butylacrylate core and a polymethacrylate shell has a greater effect on notched Izod impact strength for polyglutarimide/nylon 6, 11, and aromatic nylon compatible polymer blends (50/50) than on polyglutarimide alone.

10 Compatible polymer blends of polyglutarimide and polyamide may also exhibit improved ductility as determined by their percent elongation at break and yield as compared with polyglutarimide alone. The percent elongation at break is determined by a test conducted on an Instron tensile device (ASTM 638) using a tensile bar prepared from the material to be tested. The tensile bar is clamped at its ends in a pair of jaws of the device and the jaws are pulled apart at a uniform pre-set rate. As the tension on the tensile bar increases, the neck region of the tensile bar elongates, either stretches or yields, and eventually breaks. Yield is defined as the point at which the material begins to flow in response to stress. Yield is a function of molecular reorientation of the tensile bar in an attempt to accomodate to the tension applied. (See 1978 Book of ASTM Standards, Vol. 35, pp. 220-235). Polyglutarimide alone is brittle. It typically exhibits only 5% elongation at yield. Impact modified polyglutarimide containing a butadiene-based impact modifier, an all acrylic impact modifier, and polycarbonate exhibits a maximum percent elongation at break of 18%. Polyamide, and particularly aliphatic nylon (nylon 6 and nylon 11), is very ductile. Pure nylon 6 exhibits a 190% elongation at break while nylon 11 exhibits a 260% elongation at break.

30 It has been further found that compatible polymer blends of polyglutarimide and polyamide have improved ductility compared to polyglutarimide alone. The preferred compatible polymer blends of 50% by weight of polyglutarimide (140° methyl imide) and 50% by weight of nylon 11 exhibits a maximum percent elongation at break greater than polyglutarimide alone and about 80% higher than

he maximum % elongation at break for conventionally impact  
(polycarbonate) modified polyglutarimide. Polyglutarimide  
(ammonia imide) containing 5% styrene comonomer by weight  
5 blended with nylon 6 or aromatic nylon in a 50/50 weight  
ratio may also exhibit improved ductility compared to the  
polyglutarimide component alone.

Compatible polymer blends of low service temperature  
polyglutarimide and nylon 6 and nylon 66 exhibit an improved  
Vicat softening temperature than the corresponding  
10 polyglutarimide component alone when the nylon is present in  
the blend at concentrations of 50% or higher. Increasing  
the Vicat temperature is desirable as it permits a  
thermoplastic to be useful for applications which require  
exposure to high temperatures.

15 The following examples in which the percentages are by  
weight are presented to illustrate a few non-limiting  
embodiments of the invention.

#### General Preparation and Testing Methods

Polyglutarimide used in the following examples was  
20 prepared according to the teachings of U.S. Patent  
4,246,374. Polyamide used herein comprises nylon 6,  
manufactured by the Foster Grant Company, American Hoechst  
and BASF, nylon 11, manufactured by Rilsan Company, nylon  
66, manufactured by E.I. duPont De Nemours under the mark  
25 Zytel<sup>®</sup>, and an aromatic polyamide, Trogamid<sup>®</sup> T,  
manufactured by the Kay Fries Company.

#### Examples 1-17

##### Melt Rheology and Service Temperature Improvement of Compatible Polyamide/Polyglutarimide Blends

30 140° Vicat N-methyl and unsubstituted (ammonia)  
glutarimide polymer was prepared according to the teaching  
of U.S. Patent 4,246,374. The glutarimide polymer was

separately melt blended with nylon 6, nylon 66, and nylon 11 in a 0.8 inch Welding Engineer twin screw extruder at 525° F (barrel/die). The melt blends formed were compatible and exhibited improved melt flow rate over the corresponding polyamide component alone. The blends also exhibited Vicat softening temperature at least equivalent to or better than the corresponding polyglutarimide component alone. All the compatible polymer blends showed excellent hot strength (melt strength) while nylon 6, nylon 11, and nylon 66 alone showed poor hot strength under identical extrusion conditions and were impossible to strand and pelletize. The melt rheology and service temperature improvement of compatible polyamide/polyglutarimide blends are illustrated in Table I.

Since the melt flow rate of polyglutarimide is at least an order of magnitude slower than the melt flow rate of aliphatic nylon components alone (Example 17 versus Examples 1, 14, and 15), compatible polymer blends of aliphatic nylon and at least 5% by weight of polyglutarimide exhibit a significantly reduced melt flow rate and a correspondingly increased melt strength compared to the aliphatic nylon alone, as illustrated in Examples 15 and 16.

The Vicat softening point of polyglutarimide can be improved by incorporating aliphatic nylon in a compatible polymer blend of polyglutarimide and nylon. As illustrated by Examples 1-17, as the concentration of the imide is reduced below 80% by weight, the Vicat softening point of the compatible polymer blend becomes higher than the imide component. All compatible polymer blends of polyglutarimide and aliphatic and/or aromatic nylon containing less than 75% by weight of polyglutarimide and more than 25% by weight of nylon exhibit a Vicat softening point greater than the Vicat softening point of the corresponding imide component.

- 15 -

TABLE I

Example No.	Imide Type	Amide Type	Blend Ratio Imide/Amide	Vicat °C	Melt Flow Rate (g/10 min.)	
					450°F 400 psi	525°F 400 psi
1*	—	Nylon 6	0/100	—	195.0	478
2	140° Ammonia	Nylon 6	20/80	>200	8.5	—
3	" "	" "	50/50	172	2.3	—
4	" "	" "	80/20	135	4.8	—
5	140° Methyl	Nylon 6	20/80	199	9.1	
6	" "	" "	50/50	152	3.9	
7	" "	" "	80/20	135	10.0	
8	140° Methyl	Nylon 66	20/80	>200	—	9.0
9	" "	" "	50/50	217	—	7.5
10	" "	" "	80/20	137	2.1	34.0
11	140° Ammonia	Nylon 66	20/80	>200	—	12.0
12	" "	" "	50/50	173	—	7.2
13	" "	" "	80/20	135	2.0	42.0
14*	—	Nylon 66	0/100	—	—	83.0
15*	—	Nylon 11	0/100	—	320.0	—
16	140° Methyl	Nylon 11	5/95	—	14.0	—
17*	140° Methyl	—	100/0	140	8.2	—

\* Comparative



Examples 18-35

5           An 18 inch Welding Engineer screw extruder was employed  
to prepare a series of methyl imide/nylon 6 and methyl  
imide/nylon 11 compatible polymer blends at 470° to 525° F.  
10       The blends were injection molded using an ASTM cluster mold  
in a Newbury injection molder. The molding conditions were  
475° F barrel/nozzle, 60-70 RPM screw speed, 25 second  
injection time, and 55 second cycle time. The DTUFL, impact  
properties, ductility, modulus, and optical appearance of  
15       these blends were evaluated and the results are presented in  
Table II.

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<sup>1</sup> Imide Type is methyl.<sup>2</sup> NB = No Break

\* Comparative

TABLE II  
Impact Properties

Example No.	Imide Type <sup>1</sup>	Nylon Type	Blend Imide/Nylon	DIVUL, °C 264 psi	Izod ft/lb	GVHIT in/lb	% Elongation at Break	Modulus psi	Optical Appearance
18	140°	6	5/95	52.0	16	>340	52	306,000	Translucent
19	" "	"	50/50	99.5	8.5	>300	66	545,000	Transparent
20	" "	"	95/5	112.5	6.0	—	38	568,000	Clear
21	140°	11	50/50	67.5	NB <sup>2</sup>	>340	129	162,000	Clear
22	" "	"	95/5	108.5	6.0	—	27	526,000	Clear
23	150°	6	50/50	100.5	17	—	57	412,000	Translucent
24	" "	"	95/5	114.5	7.0	—	51	558,000	Clear
25	150°	11	50/50	73.0	NB <sup>2</sup>	>340	90	157,000	Clear
26	" "	"	95/5	111.5	7.0	—	30	551,000	Clear
27	160°	6	25/75	63.0	NB <sup>2</sup>	—	30	562,000	Translucent
28	" "	"	50/50	101.0	12.4	—	80	396,000	Translucent
29	" "	"	75/25	130.0	11.5	—	30	562,000	Transparent
30	160°	11	25/75	49.0	NB <sup>2</sup>	>340	—	—	Clear, Hazy
31	" "	"	50/50	73.0	NB <sup>2</sup>	>340	—	—	Clear
32	" "	"	75/25	106.0	8.0	—	54	456,800	Clear
33*	—	6	0/100	54.0	NB <sup>2</sup>	>300	190	125,000	Translucent
34*	—	11	0/100	38.0	NB <sup>2</sup>	>340	260	12,000	Transparent
35*	160°	—	100/0	130	7.5	4	40	600,000	Clear

These examples illustrate that compatible polymer blends of polyglutarimide and nylon exhibit improvement in service temperature of low service temperature nylon (DTUFL); improvement in impact resistance of polyglutarimide; improvement in ductility of polyglutarimide, improvements in modulus of nylon; and improvement in optical appearance of the nylon component. All the compatible polymer blends exhibited a service temperature (DTUFL) equivalent to or greater than the service temperature of the corresponding aliphatic nylon component. This service temperature improvement is the result of the higher DTUFL of the polyglutarimide as compared with the DTUFL of the nylon (compare Examples 33-35). The improvement in DTUFL service temperature of aliphatic nylon also occurs with nylon 66.

The impact resistance of polyglutarimide is also improved by forming compatible polymer blends of aliphatic nylon and polyglutarimide. This improvement occurs with compatible polymer blends containing more than 5% by weight of nylon.

All the compatible polymer blends exhibited a significantly improved modulus compared to the nylon component. This improvement in modulus is achieved by incorporating at least 5% by weight of polyglutarmide (methyl) with any aliphatic or aromatic nylon having a modulus lower than the modulus of the corresponding polyglutarimide component.

Compatible polymer blends containing at least 5% to 75% by weight of nylon 11 and methyl imide, having a Vicat service temperature of 140° C and higher, are optically clear.

The 50/50 blend of 140° Vicat methyl imide and nylon 11 (Example 21) was analyzed by Differential Scanning Calorimetry (DSC) and showed a single amorphous phase having a glass transition temperature (Tg) of 65° C. This Tg is

intermediate between the Tg of the methyl imide (138° C) and the nylon 11 (38° C) and indicates that this blend has a truly miscible amorphous phase. The 50/50 blend of 150° Vicat methyl imide and nylon 11 showed similar solubility with a single Tg at 96° C. The compatible blends of Example 20 (95 methyl imide/5 nylon 11) were molded into plaques which were optically clear with no visible haze. The plaques did not change appearance upon repeated heating and cooling in a 120° C oven. This behavior is true of all 95/5 methyl imide/nylon blends where the Vicat softening point of the methyl/imide ranges from about 140°C to about 180°C.

#### Example 36

A melt blend of equal amounts by weight of nylon 6 and nylon 11 was opaque and flexible. 25 parts of this mixture was blended with 75 parts of 140° methyl imide. A compression molded plaque of the resultant composition was hazy-clear and rigid.

#### Examples 37-45

192° ammonia imide containing 5% by weight of styrene comonomer was melt blended with nylon 6, nylon 11, nylon 66, and with alkyl-substituted aromatic nylon, Trogamid®T, in a 1" Killion extruder.

Examples 37-41 and Table III illustrate the improvement in the ductility of the compatible polymer blend as compared with the imide component alone.

Examples 42-45 illustrate that compatible blends of polyglutarimide and nylon 6, 11, and alkyl-substituted aromatic nylon show improved response to impact modification when a conventional impact modifier is incorporated into the compatible blend. The compatible melt blends of Examples 37, 38, and 39 were impact modified by extrusion melt blending 2 parts of the compatible blends with one part of

all acrylic impact modifier having a butyl acrylate core and a polymethylmethacrylate outer shell. The resulting pellets were injection molded into standard ASTM test specimens and tested. The results are illustrated in Table IV.

5

Table III

Example No.	Imide Type <sup>1</sup>	Nylon Type	Blend Imide/Nylon	% Elongation at Break	% Yield
37	192°	6	50/50	60	7
38	192°	TAT <sup>2</sup>	50/50	30	10
39	192°	66	50/50	55	--
40	192°	11	50/50	140	--
41	192°	--	100/0	4	0

10

<sup>1</sup> Imide Type is ammonia

<sup>2</sup> TAT = Trogamid<sup>®</sup> T

TABLE IV

Example No.	Imide Type <sup>1</sup>	Nylon Type	Impact Modifier	Blend Imide/Nylon/Modif.	Notched Izod ft-lb/in notch
42	192°	6	all acrylic	1/1/1	1.5
43	192°	TAT <sup>2</sup>	all acrylic	1/1/1	1.2
44	192°	11	all acrylic	1/1/1	2.0
45	192°	--	all acrylic	2/0/1	0.5

<sup>1</sup> Imide Type is ammonia

<sup>2</sup> TAT = Trogamid® T

Example 46 - Isotropy in Imide/Nylon Blends

The impact resistance (GVHIT) and directional dependence of impact resistance were evaluated using injection molded and compression molded plaques. Sample A was prepared according to Example 54 of U.S. Patent No. 4,246,374 and comprised a blend of 65 percent polyglutarimide and 35 percent MBS impact modifier. Sample B was prepared according to Example 9 of U.S. Patent No. 4,217,424 and comprised 50 percent polyglutarimide modified with 17.5 percent of butadiene-based multiple stage polymer, 17.5 percent of an all acrylic impact modifier, and 15 percent polycarbonate. The butadiene-based multiple stage polymer had a first stage which is a copolymer of butadiene and styrene, a second stage of styrene, and a final stage copolymer of methyl methacrylate, acrylonitrile, and styrene. The butadiene multiple stage polymer had the following composition:

butadiene/styrene//	styrene//	acrylonitrile/	methylmethacrylate/	styrene
73.63//	11.01//	3.83	/	3.83 /3.83

Sample C was prepared according to Example 1 (Blend A) of U.S. Patent 4,254,232. Sample C comprised 39% polyglutarimide, 41% multiple stage polymer, and 20% polycarbonate. The multiple stage polymer was an MBS multiple stage polymer having a butadiene-based first stage comprising 75% of the multiple stage polymer.

Sample D was a compatible blend of polyglutarimide and nylon 11 in a 50/50 ratio.

All the samples were injection molded to provide ASTM test plaques for GVHIT tests. The blends were also compression molded to establish the physical effects of polymeric inclusion anisotropy obtained during injection molding. The results are shown in Table V. They establish that the polycarbonate containing samples (B and C)

exhibited a significant reduction in impact resistance when compression molded (isotropic dispersion of polycarbonate) compared to injection molded (flow induced anisotropic dispersion of polycarbonate).

5           The compatible polymer blend of polyglutarimide and nylon 11 (Sample D) exhibited the same impact resistance when compression molded and injection molded, thereby establishing the absence of isotropy. The impact resistance of the injection molded sample D of this invention was  
10 substantially equivalent to or greater than the impact resistance of the injection molded samples of the conventionally impact modified polyglutarimide, while the compression molded sample D of this invention exhibited superior impact resistance compared to compression molded  
15 conventionally impact modified polyglutarimide.

Table V

	<u>Sample</u>	GVHIT (in-lb)	GVHIT (in-lb)
		<u>Injection Molded</u>	<u>Compression Molded</u>
20	A	20	20
	B	300	120
	C	120	50
	D	>260	>260



CLAIMS:

1. Polymer blend comprising 1 to 99% by weight of (a) imidized acrylic polymer containing at least 5% by weight of glutarimide units and 99 to 1% by weight of (b) polyamide.
- 5 2. Blend as claimed in claim 1 wherein said imidized acrylic polymer is derived from reaction of acrylic polymer with ammonia or methylamine.
3. Blend as claimed in claim 1 or 2 wherein said polyamide comprises nylon, preferably one or more of nylon 6, nylon  
10 11, nylon 12, nylon 66 and alkyl-substituted hexamethylene diamine terephthalate polymer.
4. Blend as claimed in any preceding claim comprising 5 to 95% by weight of polyglutarimide and 95 to 5% by weight of nylon and/or nylon-containing copolymer.
- 15 5. Blend as claimed in claim 4 wherein said polyglutarimide is derived from methylamine imidization, the blend comprising up to 95% by weight of aliphatic nylon preferably nylon 6 and/or nylon 11.
6. Blend as claimed in claim 4 or 5 comprising up to 50%  
20 by weight of polyglutarimide, wherein said nylon comprises aliphatic nylon preferably one or more of nylon 11, nylon 6 and nylon 66.
7. Blend as claimed in any one of claims 4 to 6 comprising  
25 20 to 80%, preferably 25 to 80%, by weight of aliphatic nylon, said nylon optionally comprising nylon 6 and/or nylon 66.
8. Blend as claimed in any one of claims 4 to 7 comprising  
30 at least 10% by weight, preferably at least 25% by weight of polyglutarimide, said nylon is low service temperature aliphatic nylon comprising one or more of nylon 6, nylon 11, and nylon 66, and said polyglutarimide has a higher service temperature than said aliphatic nylon.
9. Blend as claimed in any one of claims 4 to 8 comprising  
35 50% by weight of polyglutarimide, optionally having a Vicat service temperature of 140°C, and 50% by weight of nylon 11.

10. Blend as claimed in any one of claims 4 to 9 wherein  
(i) said polyglutarimide is derived from methylamine  
imidization and has a Vicat service temperature of 140°C to  
150°C, said blend comprising 5 to 75% by weight of nylon 11,  
5 or (ii) said polyglutarimide is derived from methylamine  
imidization having a Vicat service temperature of 160°C,  
said blend comprising 5 to 50% by weight of nylon 11 and 95  
to 50% of said polyglutarimide, or (iii) said  
polyglutarimide is derived from methylamine imidization and  
10 has a Vicat service temperature of 140°C to 180°C, said  
nylon comprises nylon 6 and/or nylon 11, said blend  
comprising 5% by weight of said nylon and 95% by weight of  
said polyglutarimide, or (iv) said polyglutarimide is  
derived from methylamine imidization and has a Vicat service  
15 temperature of 140°C to 180°C, nylon 6, said blend  
comprising 25 to 50% by weight nylon 6 and 75 to 50% by  
weight of polyglutarimide, or (v) said polyglutarimide is  
derived from ammonia imidization and has a Vicat service  
temperature of 192°C, said nylon comprises one or more of  
20 nylon 6, nylon 11, nylon 66, and alkyl-substituted  
hexamethylene diamine terephthalate polymer, said blend  
comprising 50% by weight of said imide and 50% by weight of  
said nylon.
11. Blend as claimed in any preceding claim of  
25 polyglutarimide derived from ammonia imidization, nylon and  
two- or three-stage, core/shell, impact modifier polymer  
optionally in equal weight ratios.
12. Blend as claimed in claim 11 wherein said impact  
modifier comprises all acrylic polymer, said polyamide  
30 preferably comprising one or more of: nylon 6, nylon 11 and  
alkyl-substituted hexamethylene diamine terephthalate  
polymer.
13. Blend as claimed in claim 10 (i), (ii) or (iii) which  
is clear.
- 35 14. Use of a blend as claimed in any preceding claim in the  
production of articles of manufacture.



Europäisches Patentamt  
European Patent Office  
Office européen des brevets

(10)

(11) Publication number:

**0 109 145  
A3**

(12)

## EUROPEAN PATENT APPLICATION

(21) Application number: 83304982.8

(51) Int. Cl.<sup>4</sup>: **C 08 L 77/10**  
**C 08 L 33/24, C 08 F 8/32**  
**C 08 L 77/00**

(22) Date of filing: 30.08.83

(30) Priority: 10.11.82 US 440672

(43) Date of publication of application:  
23.05.84 Bulletin 84/21

(88) Date of deferred publication of search report: 29.10.86

(84) Designated Contracting States:  
AT BE CH DE FR GB IT LI LU NL SE

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(64) Compatible polymer blends of imidized acrylic polymer and polyamide and their use in the production of plastic articles.

(57) Polymer blends of 1 to 99% by weight of imidized acrylic polymer, containing at least 5% by weight of glutarimide units, and 99 to about 1% by weight of polyamide are disclosed which may possess attractive physical properties and may find use in the production of plastics articles.

EP 0 109 145 A3



European Patent  
Office

# EUROPEAN SEARCH REPORT

0109145

Application number

EP 83 30 4982

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl. 3)
E	EP-A-0 094 215 (E.I. DU PONT DE NEMOURS)	1-14	C 08 L 77/10 C 08 L 33/24 C 08 F 8/32 C 08 L 77/00
D, A	US-A-4 246 374 (KOPCHIK)		
A	US-A-4 255 322 (KOPCHIK)		
D, A	US-A-4 321 336 (MEYER et al.)		
			TECHNICAL FIELDS SEARCHED (Int. Cl. 3)
			C 08 L 77/00 C 08 L 33/00
The present search report has been drawn up for all claims			
Place of search BERLIN		Date of completion of the search 10-07-1986	Examiner KRAIL
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EPO Form 1503 (03/82)